

## **REMARKS**

Claims 1-19 and 22-27 stand rejected in the subject patent application, while claims 20 and 21 merely were objected to as depending from a rejected claim. Claims 1-17 remain pending after entry of this amendment.

### **Rejection Under 35 U.S.C. §112**

Claims 6-16 were rejected under the second paragraph of 35 U.S.C. §112 because the phrase “the two-dimensional array” in claim 6 lacked an antecedent basis. That phrase has been amended to “the first array” which was used previously in the claim. Thus claims 6-16 now comply with the requirements of 35 U.S.C. §112.

### **Rejection Under 35 U.S.C. §103**

Claims 1, 2, 5 and 17 were rejected under 35 U.S.C. §103 as being unpatentable over Petroff in view of the Sumitomo Japanese patent. Because the Sumitomo reference is in Japanese, applicant encloses the English language abstract published by the Japanese Patent Office which contains a further description of that invention.

The rejection cites the Petroff patent as disclosing a radiation detector for an medical imaging system that has a scintillator 10 which converts gamma ray photons 24 into visible light photons 26. The visible light photons 26 are reflected by the faces within the scintillator into two fluorescent fibers 12 and 14 running orthogonally across the radiation detector. The entering visible light photons 26 cause the fibers to fluoresce

thereby producing light 28 at a different wavelength which travels through the fibers to photon counters 20. A signal processor analyzes the photon counts to produce an image.

In contrast, the Sumitomo reference does not relate to imaging and does not have a scintillator. This apparatus analyzes liquids that have chemiluminescence, i.e. fluoresce or scintillate when exposed to ionizing radiation. With reference to Figure 3 of Sumitomo, as the liquid being assayed flows through tubes 15, the light emitted by that liquid enters an optical fiber 13. Several optical fibers 33 in Figure 5 lead from separate tubes 15 to an optical switch 36 which selectively directs light from the fibers to detection equipment 39 so that different liquids in those tubes 15 can be analyzed. For an English language paper generally explaining this technique for analyzing liquids, see Alan Hogg, *Liquid Scintillation Counting*, Radiocarbon Laboratory, University of Waikato, New Zealand, 1999, copy enclosed.

It is respectfully submitted that one skilled in the art of medical imaging, such as Mr. Petroff, would not find it obvious to investigate chemical assaying equipment, such as the Sumitomo apparatus, for devices to use in a medical imager that responds to gamma radiation. Furthermore, to support a conclusion that a claimed combination is obvious, the references must either impliedly or expressly suggest the selection of the various elements in that combination, *In re Newell*, 891 F.2d 899, 13 U.S.P.Q.2d 1248, 1250 (Fed. Cir. 1989). Nothing in the Petroff and Sumitomo Japanese patents remotely suggests how their teachings could be combined, much less combined to yield the subject matter claimed in the present application. Any contention to the contrary clearly is made

with the benefit of the description in the subject application and such use of hindsight is proscribed, *W.L. Gore & Assoc., Inc. v. Garlock, Inc.*, 712 F2d 1540, 220 U.S.P.Q. 303 (Fed. Cir. 1983).

Furthermore, the ability of an optical switch as in Sumitomo to function adequately and fast enough for assaying a plurality of chemiluminescence liquids does not indicate that an optical switch would be suitable for the detector array of a medical imaging system.

Therefore, claims 1, 2, 5, and 17 are patentable under 35 U.S.C. §103.

Claims 3, 4, 18, 19 and 22-27 have been rejected under 35 U.S.C. §103 as being unpatentable over Petroff and Sumitomo, and further in view of Judy *et al.* The latter reference was cited as teaching microelectromechanical mirrors.

However the apparatus in Judy *et al.* is used to switch light among optical fibers, as shown in Figure 4. While that might suggest using microelectromechanical mirrors in the optical switch of the Sumitomo apparatus, Judy *et al.* fails to suggest modifying the Petroff photodetector array which does not employ an optical switch. Therefore, the combination of the teachings in all three references still does not suggest the basic structure of the presently claimed invention.

Furthermore, claims 3, 23 and 24 recite a linear array and a two-dimensional array of microelectromechanical mirrors, whereas Judy *et al.* teaches only a two-dimensional array, see Figure 4. It also appears that using microelectromechanical mirrors as the optical switch Sumitomo would use only a single array as well.

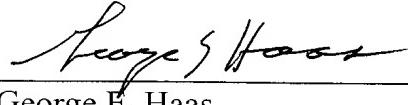
Therefore, claims 3, 4, 18, 19 and 22-27 are patentable under 35 U.S.C. §103.

## **Conclusion**

In view of these distinctions between the subject matter of the present claims and teachings of the cited patents, reconsideration and allowance of the present application are requested.

Respectfully submitted,  
Richard J. Ruzga

Dated: September 12, 2003

By:   
George E. Haas  
Registration No. 27,642

Quarles & Brady LLP  
411 E. Wisconsin Avenue Suite 2040  
Milwaukee, WI 53202-4497  
Telephone (414) 277-5751

QBMKE\5467234.1



(19)

(11) Publication number: **2001337083 A**

Generated Document.

## PATENT ABSTRACTS OF JAPAN

(21) Application number: **2000158134**

(51) Int'l. Cl.: **G01N 31/20 G01N 21/27 G01N 21/64 G01N 21/77**

(22) Application date: **29.05.00**

(30) Priority:

(43) Date of application  
publication: **07.12.01**

(84) Designated contracting  
states:

(71) Applicant: **SUMITOMO ELECTRIC IND LTD**

(72) Inventor: **HIRATA YOSHIHIRO**

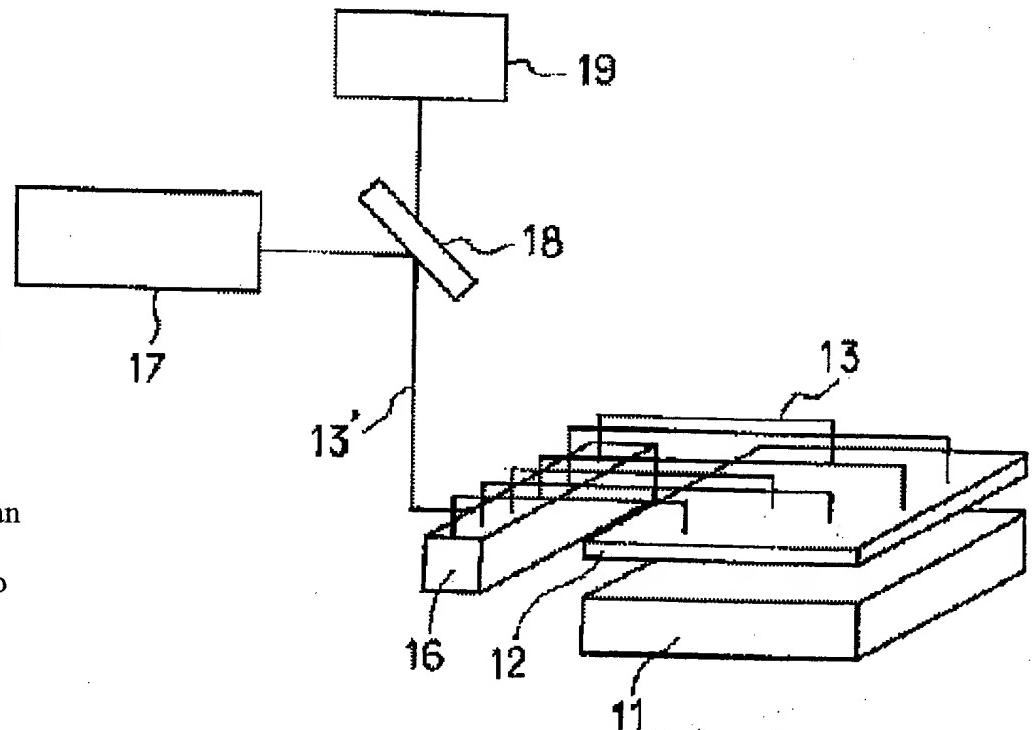
(74) Representative:

### (54) OPTICAL SYSTEM FOR MICROCHEMICAL ANALYTICAL SYSTEM

#### (57) Abstract:

**PROBLEM TO BE SOLVED:** To provide a high general purpose optical system suitable for a high-performance process for a microchemical analytical system.

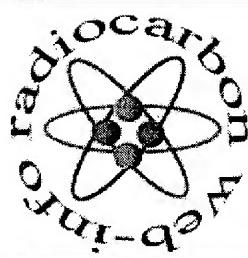
**SOLUTION:** The optical system for guiding a light from a plurality of parts to be detected (flow cells, capillaries or the like) existing in a micro-fluid chip to a sensor 19 comprises an optical switch 16 and an optical fiber 13 connected to the switch 16. The fiber 13 is opposed to the respective parts of the chip 11. The switch 16 selects a necessary optical path by a switching mechanism as needed.



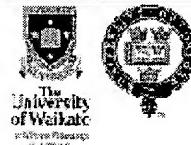
COPYRIGHT: (C)2001,JPO



from the site..



- The method
- Measurement
- Applications
- WWW links
- K-12
- Publication
- Corrections
- Age calculation
- Calibration
- Pretreatment
- Bibliography
- Awards
- Credits
- What's New
- Email



This WWW site is copyright, may not be reproduced without permission.

Welcome to radiocarbon WEB-info. Radiocarbon dating is the technique upon which chronologies of the late Pleistocene and Holocene have been built. This resource is designed to provide online information concerning the radiocarbon dating method. We hope it will be of occasional use to radiocarbon users and interested students alike.

Copyright © 1999 Tom Higham, Radiocarbon Laboratory, University of Waikato, New Zealand (Email: [thomas.higham@archaeology-research.oxford.ac.uk](mailto:thomas.higham@archaeology-research.oxford.ac.uk)).



[INDEX](#) | [Introduction](#) | [Measurement](#) | [Applications](#) | [WWW Links](#) | [K12](#) | [Publication](#) | [Corrections](#) |  
[Age calculation](#) | [Calibration](#) | [Pretreatment](#) | [References](#) | [Awards](#) | [Credits](#) | [What's new?](#) | [Email](#) |

LAST MODIFIED 16 May 2002  
HTML DOCUMENT BY [T. HIGHAM](#).

---

# Liquid Scintillation Counting (LSC)

---

CONTRIBUTED BY: ALAN HOGG

**DISCLAIMER:** The methods described below are not universally applied in all LSC radiocarbon laboratories, there may be slight variations in some of the procedures between different laboratories and radiocarbon specialists. For detailed information regarding the pretreatment procedures employed by a specific laboratory it is necessary to contact the laboratory in question. Data given below is merely intended as a guide only.

---

## Introduction

The method of  $^{14}\text{C}$  counting used by Libby and his co-workers involved measuring radioactivity using modified Geiger counters. The next development in counting technology was the conversion of sample carbon to  $\text{CO}_2$  gas for measurement in Gas Proportional counters. In the early 1950's, the first attempts were made to detect  $^{14}\text{C}$  by the Liquid Scintillation (LS) counting method. In the 1940's, Broser and Kallman (1947) discovered that certain organic compounds (scintillators) fluoresced when exposed to ionising radiation. Each fluorescence event is proportional to a radioactive decay event, and the frequency of these events is directly proportional to the number of  $^{14}\text{C}$  atoms present in the sample. A brief summary of the LS method and some of the problems associated with the technique are given here. A more detailed report can be found in Polach (1987). Discussion on the LS method can be broken down into a number of topics : counting liquid, counting vials, and the LSC method, including counting instrumentation and potential problems associated with measurement of beta ( $\beta$ )-decay by LS counting.

## Counting Liquid

In the majority of LSC facilities, the scintillation solvent is benzene ( $\text{C}_6\text{H}_6$ ) or a mixture of benzene and toluene ( $\text{C}_6\text{H}_5\text{CH}_3$ ). Benzene has been chosen because of its excellent light transmission properties and the high chemical conversion yield of sample C to benzene. The sample is first converted to  $\text{CO}_2$ , then reacted with molten lithium to form lithium carbide ( $\text{Li}_2\text{C}_2$ ), before being catalytically trimerised to benzene. Benzene synthesis procedures vary considerably in different laboratories. The details outlined below describe the procedures used in the Waikato laboratory to give the reader some idea of how the chemical conversions may be carried out.

The carbon is first oxidised to  $\text{CO}_2$ , either by acid hydrolysis (for carbonates), or combustion in an oxygen stream or combustion bomb (for organic materials). The combustion gases are passed over heated  $\text{CuO}$  to complete the oxidation of  $\text{CO}$ ,  $\text{NO}$ ,  $\text{N}_2\text{O}$  and  $\text{NO}_2$  and also of tar substances. The  $\text{CO}_2$  may then be then purified using a chain of wet chemical reagents; for example,  $\text{AgNO}_3$  and  $\text{Hg}(\text{NO}_3)_2$  to precipitate halogens, and the oxidising agents  $\text{KI}/\text{I}_2$  and  $\text{K}_2\text{Cr}_2\text{O}_7$  to remove nitrogen and sulphur compounds. Silica gel and dry ice traps ( $-80^\circ\text{C}$ ) remove any water remaining in the gas (see figure 1).

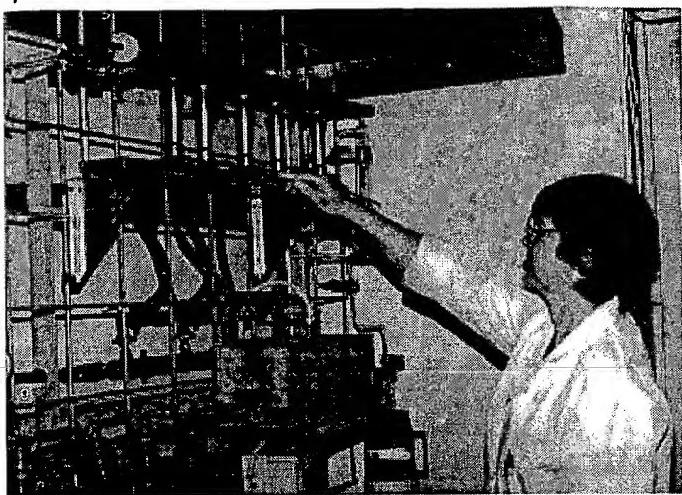
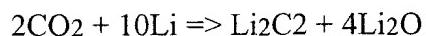
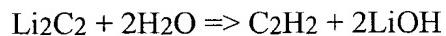


Figure 1: Combustion system for preparation of CO<sub>2</sub> from organic carbon materials (see text for details).

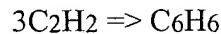
The purified CO<sub>2</sub> is then reacted with molten lithium in a stainless steel or inconel reaction vessel in vacuo:



This reaction was first described by Barker (1953), who developed the reaction using lithium instead of barium, and later improved by Polach and Stipp (1967). The CO<sub>2</sub> is bled slowly onto the molten Li where it is converted to Li<sub>2</sub>C<sub>2</sub>. The carbide is heated to ca. 800°C (furnace temperature) and placed under active vacuum for 30 minutes to remove any unreacted gases and complete the carbide synthesis (Gupta and Polach, 1985). The lithium carbide is cooled and then hydrolysed to acetylene gas (Li<sub>2</sub>C<sub>2</sub>):



The acetylene is purified by passing through a phosphoric acid trap to remove ammonia compounds, and again, dry ice traps to remove water vapour. Finally the acetylene is trimerised to benzene using a suitable catalyst. There are a variety of vanadium or chromium activated catalysts available, including a silica-alumina vanadium activated catalyst developed by John Noakes (CAIS, Univ. of Georgia), a vanadium-alumina-silica catalyst produced at the Institute of Geography, Univ. of Petersburg by Dr Kh. A. Arslanov and another at the British Museum by Dr J. Ambers. The catalytic trimerisation for some catalysts may be more efficient at reduced temperatures (e.g. 5°C).



Benzene is then driven off the catalyst at ca. 100°C and collected under vacuum at ca. -65°C. The benzene is then stored in a vial under refrigeration to await counting.

This sequence of reactions requires a high degree of operator skill because of the complexity of the equipment and the nature of the reactions. It is important that a standardised routine is followed carefully and consistently, so that yields remain high and there is little cross-contamination between samples.

### Counting vials

The synthesised benzene is transferred into counting vials of a variety of types. The counting vials contain the sample solvent and the scintillator. Commercially available LS vials used for benzene counting are commonly composed of either Teflon, quartz or low-K glass (e.g. Polach et al. 1983; Haas 1979; Devine & Haas 1987; Noakes & Valenta 1989; Hogg & Noakes, 1992; Hogg 1993). Polyethylene and polypropylene vials are very useful for non-aromatic solvents, but their permeability makes them unsuitable for repeatable long-term benzene use. In a comparison of different vial types, Hogg et al. (1991) concluded that, for spectrometers using true anticoincidence detection and extensive passive shielding (as in the Wallac 1220 Quantulus), Teflon or synthetic silica vials produced the highest performance in terms

of  $^{14}\text{C}$  detection efficiency and background. The authors also noted that synthetic silica had superior physical properties to Teflon. The silica vials vary in size according to the weight of the sample benzene being counted, including 0.3 ml and 1 ml minivials, 3 ml standard vials and 12 ml vials, suitable for high precision applications. Low-K glass vials are supplied in either 7 ml or 20 ml sizes.

The most commonly used scintillator in  $^{14}\text{C}$  dating is PPO + POPOP, either dissolved in toluene or directly in sample benzene. Polach et al (1983) made a comparative study of various scintillators and concluded that dry powder butyl-PBD dissolved in sample benzene (15g/l) gave superior and stable performance even under extreme quench conditions. Some Packard spectrometers utilise an active plastic holder (Pico adaptor) which helps to reduce background radiation. These instruments require a secondary wave shifter (usually bis-MSB, see Cook, Harkness & Anderson 1989) to be added to the butyl-PBD. In the Waikato laboratory, the scintillator (butyl-PBD) is dissolved directly into the sample benzene at a concentration of 15g/l. The vial is then transferred into a Quantulus spectrometer and allowed to cool and dark adapt for a minimum of 8 hours prior to the commencement of counting.

### **Liquid Scintillation Spectrometry**

The essential electronic components of the Liquid Scintillation (LS) Spectrometer according to Gupta and Polach (1985), are photo multiplier tubes (PMTs), high voltage supply, signal preamplifiers, pulse and summing amplifiers, coincidence logic, timer and scaler (Gupta and Polach, 1985:50). The LS spectrometer measures electronically the pulses of light generated from photon emissions emitted by a scintillator in response to a radioactive decay event. The PMTs register an electronic pulse proportional to the energy of a particular beta decay when a photon or light particle is emitted within the benzene cocktail. The anode current generated from a PMT, then, is a function of the level of radioactivity (Horrocks, 1974).

### **External factors interfering with $\beta$ -decay detection by LSC**

#### **Optical cross talk**

Optical cross-talk occurs between opposed PMTs. An event occurring in one tube initiates a pulse in the other (Noakes, 1977). Ionising events may occur in either PMT due to interactions between the molecules of the tube material and surrounding radiation. Butterfield and Polach (1983) and Gupta and Polach (1985) have described methods used to reduce optical cross talk in LS counters, both by optimising and refining electronic circuitry to enable differentiation between sample and non-sample events and by masking areas in the counting chamber to reduce reflection. Carefully designed vials and vial holders can also help to reduce cross-talk, by minimising the view of opposing PMTs to each other.

#### **Line transmission noise**

Line transmitted switching noises and variation in the high voltage supply can influence reproducibility in LS counting. Power supplies are therefore frequently filtered and regulated, either with line noise suppression electronic transformers or a suitable uninterruptable power supply (UPS) unit. A clean and stable voltage supply is crucial because of the relationship between electronic particle detection and the energy of the decay event.

#### **Radiofrequency interference**

Radiofrequency interference is generated by lights, motors and switches in the vicinity of the counter. RF detection circuitry can be fitted which will identify RF signals and ensure they are removed from the sample  $^{14}\text{C}$  spectrum.

#### **Static induced noise**

Static induced noise results from the build up of static electricity through friction generated from the movement of vials throughout the counting system. Earthing of vials and holders and an ionising unit which the vials move past, can be used to remove static electricity prior to counting.

#### **Radon contamination**

<sup>222</sup>Rn emits alpha and beta particles, the latter with a decay energy of 5.587 meV. This decay interferes with the <sup>14</sup>C spectrum, resulting in higher count rates and radiocarbon ages that are too young. Radon's short half-life of 3.82 days, however, means that a delay in counting eliminates the problem. Sample benzene is therefore often left for 3 - 4 weeks to allow any radon that may be present to decay. Potential <sup>222</sup>Rn contamination is more significant in older samples because of their lower count rates.

### Natural radioactivity and cosmic radiation

Natural atmospheric radiation influences sample count rates. This is reduced through shielding of the counting environment. This may be passive or active shielding. The passive shield consists of lead which shields the counting chamber from external radiation. The term 'active shielding' describes electronic means of recognising and eliminating background causing events from the sample beta spectrum. Active shielding occurs in two forms in modern LS counters. The Wallac Quantulus, utilises an electronic anti-coincidence guard, comprising a liquid scintillator guard containing its own phototubes, surrounding the counting chamber and sample PMTs. Ionising radiation from non-sample external sources passing through the guard scintillator leaves excited atoms and molecules in it which are seen by the guard PMTs. If an ionising event is recorded simultaneously in the guard and sample tubes, it is rejected as a background count. Some Packard counters utilise 'time resolved counting' to differentiate between, sample events with a low pulse index (see Roessler et al, 1991 for details), and background events with a higher pulse index.

Most higher performance counters use multi-channel analysers (MCAs) to store and evaluate counting data. The MCAs are software-controlled to provide complete flexibility in data acquisition.



LSC Dating by the Wallac Oy "Quantulus"

### Summary

One of the major advantages of LSC is that sample, background and modern vials are able to be rotated into the counting chamber by the operator. This is termed "quasi-simultaneous counting" and was first implemented by Noakes, Kim and Stipp (1965). Major developments in LSC in the last decade include the optimisation of spectrometers, incorporating sophisticated computer and electronic hardware which enables the user to validate counting data with ease. In addition, background count rates have been significantly reduced through the addition of active and passive forms of shielding, including lead and through the placement of LS spectrometers underground. A number of researchers have participated in the development of 'minivial' technology which has enabled samples of <1 g carbon to be dated through the LSC method.

One of the most widely known applications of LSC of C14 is in high precision dating of the radiocarbon timescale for the purposes of calibration. This was conducted primarily at the Belfast Radiocarbon Dating Laboratory (an LSC facility) and the Quaternary Dating Centre at the University of Washington (Gas Proportional Counting). The following LSC laboratories present information on the technique and its application:

- University of Waikato Radiocarbon Dating Laboratory (New Zealand).
- Beta Analytic Radiocarbon Dating Laboratory (USA)
- Queen's University of Belfast - Radiocarbon Laboratory (UK)

For a review of LSC see: Polach, H.A. (1992) Four Decades of LS Counting and spectrometry. in, *Radiocarbon after four Decades. An interdisciplinary Perspective.* (eds) Taylor, R.E, Long, A and Kra, R. Springer-Verlag, NY.

Nevada Technical Associates, Inc. has some web pages devoted to radiation safety, radiochemistry and related areas. Equipment including Liquid Scintillation Counters is available here.

Another manufacturer of LSC technology is Packard.